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Annually resolved $\delta^{13}C_{shell}$ chronologies of long-lived bivalve mollusks (*Arctica islandica*) reveal oceanic carbon dynamics in the temperate North Atlantic during recent centuries

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ABSTRACT

The ability of the ocean to absorb carbon dioxide is likely to be adversely affected by recent climate change. However, relatively little is known about the spatiotemporal variability in the oceanic carbon cycle due to the lack of long-term, high-resolution dissolved inorganic carbon isotope ($\delta^{13}C_{DIC}$) data, especially for the temperate North Atlantic, which is the major oceanic sink for anthropogenic CO₂. Here, we report shell carbon isotope values ($\delta^{13}C_{shell}$), a potential proxy for $\delta^{13}C_{DIC}$, of old-grown specimens of the long-lived bivalve mollusk, *Arctica islandica*. This paper presents the first absolutely dated, annually resolved $\delta^{13}C_{shell}$ record from surface waters of the North Atlantic (Iceland, Gulf of Maine) covering the time interval between 1753 and 2003. According to our results, the $\delta^{13}C_{shell}$ data were unaffected by trends related to ontogenetic age. However, the shell carbonate was precipitated with a constant offset from expected equilibrium by -1.54 to $-2.7 \pm 0.2\%$ corresponding to a 6.2 to $10.8 \pm 0.8\%$ contribution of respiratory CO₂ (-25%). The offset did not appear to vary through the lifetime of individual specimens and among specimens. Therefore, the $\delta^{13}C_{shell}$ data of this species can very likely be used as a measure of $\delta^{13}C_{DIC}$.

Furthermore, shell stable carbon isotope chronologies exhibited habitat-specific differences and a significant inter-annual and decadal variability related to the natural carbon cycle. In addition, a distinct negative $\delta^{13}C_{\text{shell}}$ shift was found reflecting the oceanic Suess effect, i.e. the admixture of anthropogenic CO₂. However, this shift only occurred after the early 1920s when a major climate regime shift led to a northward movement of the oceanic Polar Front in the Nordic Seas and a large-scale reorganization of atmospheric and oceanic currents in the North Atlantic. This likely resulted in a reduced admixture of cold Polar water onto the North Icelandic shelf (through the East Iceland Current) and the Gulf of Maine (through the Labrador Current) with an increased volume of warmer, isotopically well-equilibrated Atlantic waters. Our shell-based $\delta^{13}C_{\text{DIC}}$ proxy record provides the basis to quantitatively assess natural and anthropogenically induced patterns of carbon uptake in the North Atlantic.

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1. Introduction

Approximately $118 \pm 19 \text{ Pg C}$ ($1 \text{ Pg C} = 10^{15} \text{ g C} = 1 \text{ Gt carbon}$), i.e. ca. 30% of the total carbon dioxide emitted to the atmosphere by human activity between 1800 and 1994 has been absorbed by the oceans (Sabine et al., 2004) in the form of dissolved inorganic carbon (DIC). Carbon dioxide (CO₂) resulting from the combustion of fossil energy sources, deforestation and changes in land use is depleted in

¹³C (Tans, 1981). During the last 200 years the stable carbon isotope ratio of atmospheric CO₂ ($δ^{13}C_{atm}$) has thus changed from values of ca. - 6.3 to - 8.1‰ (Friedli et al., 1986; Francey et al., 1999; Keeling et al., 2005). Furthermore, the admixture of anthropogenic CO₂ has led to a significant negative shift of the stable carbon isotope ratio of the oceanic DIC ($\delta^{13}C_{DIC}$) (Nozaki et al., 1978; Druffel and Benavides, 1986; Böhm et al., 1996; Swart et al., 1998; Lazareth et al., 2000; Moore et al., 2000; Böhm et al., 2002), also known as the oceanic Suess effect (Gruber et al., 1999). The rate of change of the $\delta^{13}C_{DIC}$ signature can be used to estimate the oceanic uptake rate of atmospheric CO₂ (Gruber et al., 2002; Quay et al., 2003, 2007). This uptake rate, however, underlies significant spatiotemporal variations which are poorly

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